



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Effect of fluoride in NTS groundwaters on the aqueous speciation of U, Np, Pu, Am and Eu

Carol J. Bruton, Gregory J. Nimz

March 24, 2005

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

Effect of fluoride in NTS groundwaters on the aqueous speciation of U, Np, Pu, Am and Eu

Carol J. Bruton and Gregory J. Nimz
Lawrence Livermore National Laboratory

Summary: To address SNJV concerns that fluoride in Nevada Test site (NTS) groundwaters may impact radionuclide speciation and transport, NTS water quality databases were obtained and scanned for analyses with high fluoride concentrations (> 10 mg/L). The aqueous speciation of nine representative samples of these groundwaters with added trace amounts of uranium (U), neptunium (Np), plutonium (Pu), americium (Am) and europium (Eu) was then calculated with the computer code EQ3NR assuming a temperature of 25°C , using currently available thermodynamic data for these species. Under conditions where U(VI), Np(V), Pu(IV), Am(III) and Eu(III) dominate, F complexes are insignificant (< 1 mole %) for U, Np, Pu and Am. Eu-F complexes may be significant in groundwaters that lack bicarbonate, possess pH values less than about 7 at ambient temperatures, or contain F in extremely high concentrations (e.g. > 50 mg/L).

Objective: Evaluate the extent to which fluoride in NTS groundwaters complex U(VI), Np(V), Pu(IV), Am(III) and Eu(III).

Approach: Screen existing databases of groundwater chemistry at NTS for waters with high fluoride concentrations and calculate the extent to which fluoride complexes with the nuclides of interest in these waters.

Fluid chemistry: Databases containing fluid chemistry of NTS waters were obtained from Stoller-Navarro (2004), Rose (2005) and SNJV (2005). The Stoller-Navarro database is the primary Underground Testing Area (UGTA) database. Figure 1 shows all of the F analyses in this UGTA database. Of the 2991 F analyses in the database, 66 have F greater than 10 mg/L. These 66 analyses are from 34 different wells and springs, 24 of which are external to the NTS boundary (38 F analyses) and 10 are wells within the NTS (28 F analyses). The Rose (2005) database contains 8 analyses for 6 NTS wells that have F greater than ~ 10 mg/L. The SNJV (2005) database contains 11 analyses for 6 NTS wells with F greater than 10 mg/L. All of the wells in the SNJV (2005) database, and all but one of the wells in the Rose database, are also in the UGTA database, although the analyses from these databases that are used in this report have not yet been entered into the UGTA database. The resulting 11 NTS wells producing ground water with F concentrations greater than ~ 10 mg/L are:

NC-EWDP-1DX (deep)
U-20n PS#1 DDH
U-19q PS#1d
ER-20-5 #1
ER-5-4 #2
ER-5-4
ER-18-2
U-19as (3584 ft)

U-19bh
UE-19c Water Well
U-2bs PS #1DB (Starwort)

Of these wells, 9 were chosen for evaluation on the basis of representative water chemistry, range of F concentrations (9 to 80 mg/L), geographical location, and date of sampling. Chemical analyses for all NTS waters with F greater than 10 mg/L are given in Table 1, with the 9 analyses chosen for aqueous speciation evaluation highlighted.

Speciation Evaluation Assumptions: The temperature of all waters was assumed to be 25°C, even though temperatures, when reported, are higher (about 33-43°C). Temperature-dependent thermodynamic data for radionuclide complexes are generally not available. The small temperature differences should not significantly alter the findings of this study. Total carbonate was used when reported or, if not, HCO_3^- .

To calculate the speciation of U, Np, Pu, Am and Eu in these waters, we assumed radionuclide concentrations of 10^{-12} M. In evaluating the degree to which the radionuclides are complexed with available ligands (e.g. OH^- , CO_3^{2-} , F) and the competition of the ligands for the radionuclide, the absolute concentration of the radionuclide chosen is arbitrary (unless precipitation of radionuclides as solid phases is being considered). Note that naturally occurring U and Eu will likely be present in NTS groundwaters that will compete for F with their radioactive counterparts. However, these natural concentrations are not considered in this study.

An oxygen fugacity of 10^{-30} bars was assumed. At this oxygen fugacity and the measured pH values of the groundwaters, the dominant oxidation states of the radionuclides studied were as follows: U(VI), Np(V), Pu(IV), Am(III) and Eu(III). These oxidation states are currently thought to be relevant to most environmental conditions along potential flow paths from test cavities in the saturated zone at NTS.

Computer codes and thermodynamic data used: The LLNL EQ3NR speciation-solubility code with the data0.ymp.R4 thermodynamic database (Wolery, 2002) was used to make the speciation calculations. The data0.ymp.R4 thermodynamic database contains Nuclear Energy Agency data for the radionuclides of interest from Grenthe et al. (1992), Lemire, (2001), Silva et al. (1995) and Spahiu and Bruno (1995). Table 2 lists the F species in the data base that complex U, Np, Pu, Am and Eu and other components of the selected groundwaters. The aqueous species $\text{Pu}(\text{OH})_4(\text{aq})$, which is considered to be a major Pu aqueous species, was not included in this data base, probably because the NEA did not consider available data to be sufficiently accurate. Nonetheless, $\text{Pu}(\text{OH})_4(\text{aq})$ is recognized as an important species and it is better in this case to include even uncertain data than to omit it entirely. The data for $\text{Pu}(\text{OH})_4(\text{aq})$ is available from other EQ3NR data bases, namely data0.cmp.R2. Data for $\text{Pu}(\text{OH})_4(\text{aq})$ from Lemire and Tremaine (1980) as contained in data0.cmp.R2 was therefore added to the data0.ymp.R4 data base.

Results: Tables 3 through 7 and Figures 2 through 6 show the results of the speciation calculations in terms of the percentages of the total concentrations of U, Np, Pu, Am and Eu in solution that are complexed by a given F species. Results are expressed in mole percent. Concentrations of HCO_3^- (total carbonate as reported or bicarbonate) and F are also shown in mg/L because of their relevance in determining the aqueous speciation.

Uranium - U-F complexes do not account for a significant percentage (generally much less than 0.01%) of the total aqueous U in solution (Table 3, Figure 2). The U(VI) $\text{UO}_2(\text{CO}_3)_3^{4-}$ and $\text{UO}_2(\text{CO}_3)_2^{2-}$ species generally comprise > 98% of the total U in solution.

Neptunium – Np-F complexes account for a maximum of about 1% of the total aqueous Np in solution (Table 4, Figure 3). The Np(V) complex $\text{NpO}_2\text{F}(\text{aq})$ complex dominates the Np-F species. The Np(V) complexes $\text{NpO}_2\text{CO}_3^-$ and NpO_2^+ generally comprise > 98% of the total Np in solution.

Plutonium – Pu-F complexes do not account for a significant percentage (generally much less than 10⁻⁷%) of the total aqueous Pu in solution (Table 5, Figure 4). The Pu(IV) complex $\text{Pu}(\text{OH})_4(\text{aq})$ generally comprises > 97% of the total Pu in solution.

Americium - Am-F complexes account for a maximum of 1% of the total aqueous Am in solution (Table 6, Figure 5). The Am(III) species $\text{Am}(\text{CO}_3)^+$, $\text{Am}(\text{CO}_3)_2^-$, $\text{Am}(\text{CO}_3)_3^{3-}$, $\text{Am}(\text{OH})_2^+$ and $\text{Am}(\text{OH})^{2+}$ generally comprise > 98% of the total Am in solution.

Europium - The dominance of Eu-F species is variable. The presence of bicarbonate in solution and the pH affects the complexation of Eu by F (Table 7, Figure 6). In sample ER-20-5#1, Eu-F complexes comprise about 60% of the total Eu in solution because HCO_3^- is not reported (Table 1) and is not considered in the EQ3 evaluation. Eu(III)- CO_3 species ($\text{Eu}(\text{CO}_3)_2^-$, EuCO_3^+) generally dominate Eu complexation, and in its absence, Eu-F complexes form. Sample ER-20-5#1 has a major charge imbalance, which probably reflects that HCO_3^- was actually present, but not analyzed. This evaluation shows the potential for formation of Eu-F complexes in any waters that are HCO_3^- -depleted. However, it is important to note that, in reality, Eu-F species do not dominate Eu speciation in this water.

Eu-F complexes comprise about 5-10% of total aqueous Eu in samples UE-19c and U-19qPS1d. This is largely due to the fact that these samples have lower pH values (7.2) than the other samples (7.9 to 9.3). At a pH of 7.2, the concentration of CO_3^{2-} is very low, and EuCO_3^+ replaces $\text{Eu}(\text{CO}_3)_2^-$ as the major contributor to Eu speciation, allowing F complexes to increase in contribution. This suggests that Eu-F complexes might be significant in groundwaters with pH values less than about 7. In contrast, at pH values close to 9 and above (e.g. U-19vPS1ds), the species $\text{Eu}(\text{CO}_3)_2^-$ greatly dominates Eu speciation owing to the dominance of CO_3^{2-} in solution.

Eu-F complexes comprise about 15% of the total Eu in solution in sample U-19as, largely due to the extremely high F concentration in solution (80 mg/L).

Conclusions: Aqueous complexes of U(VI), Np(V), Pu(IV) and Am(III) with F will not comprise significant percentages of the total aqueous concentrations of these elements in NTS groundwaters at 25°C, based on the thermodynamic data and the range of representative NTS fluid chemistries chosen for this study. Aqueous complexes of Eu(III) with F may become significant in bicarbonate-free waters, in waters with pH values of about 7 or less, and in waters with extremely high F concentrations, such as 80 mg/L.

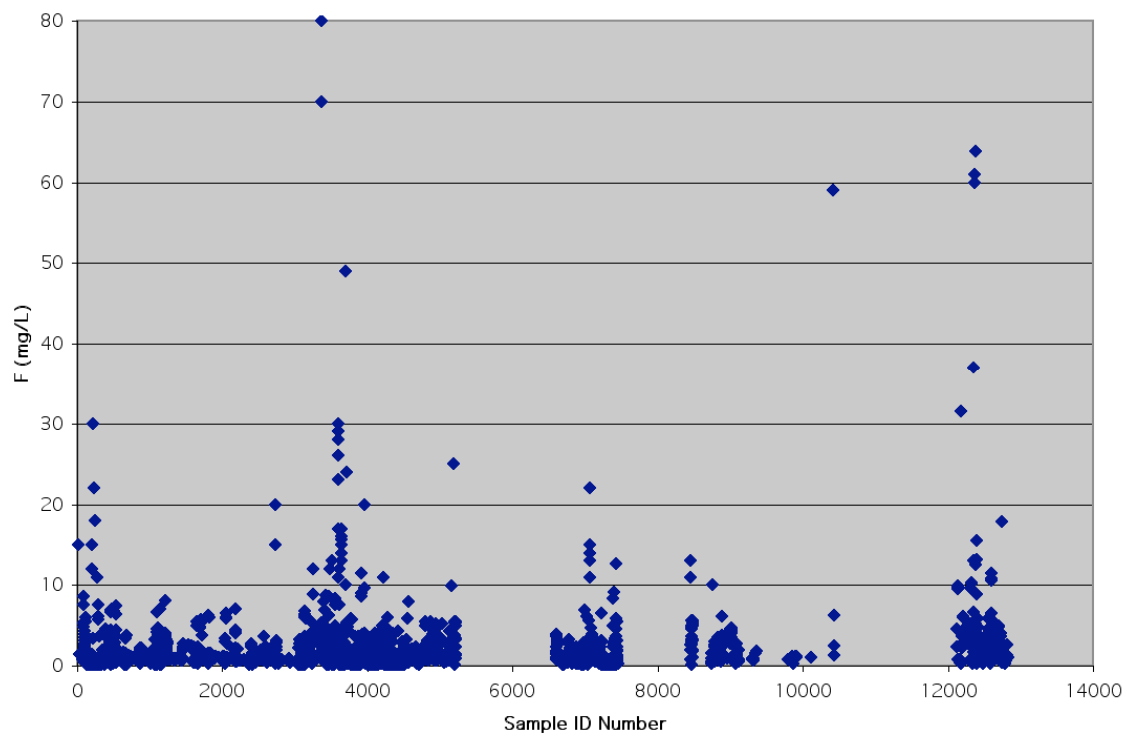


Figure 1. Fluoride concentrations in waters from the UGTA database (Stoller-Navarro, 2004).

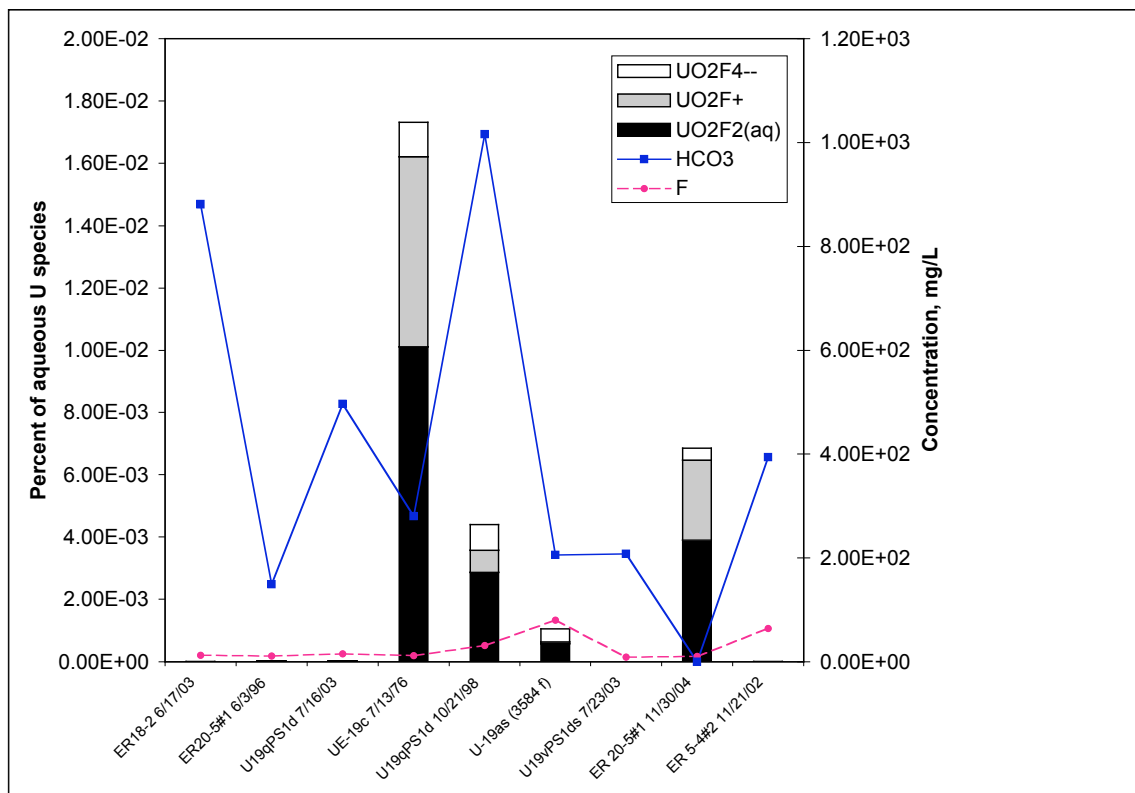


Figure 2. Aqueous uranium-fluoride species, and HCO_3 and F concentrations in nine selected NTS wells.

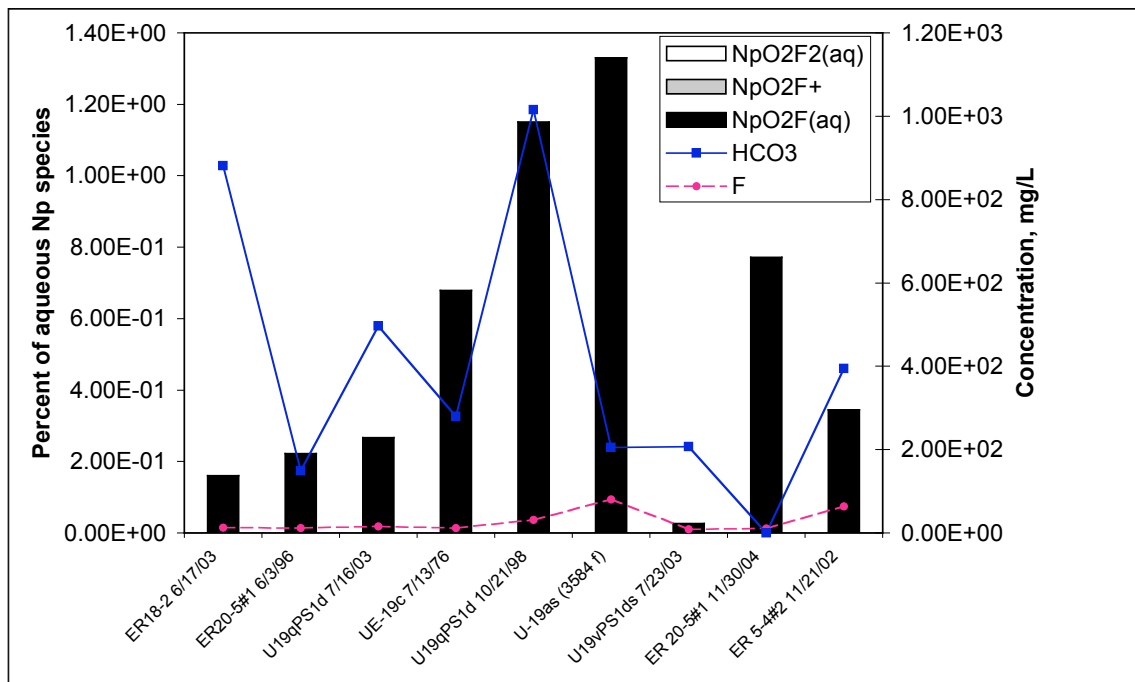


Figure 3. Aqueous neptunium-fluoride species, and HCO_3 and F concentrations in nine selected NTS wells.

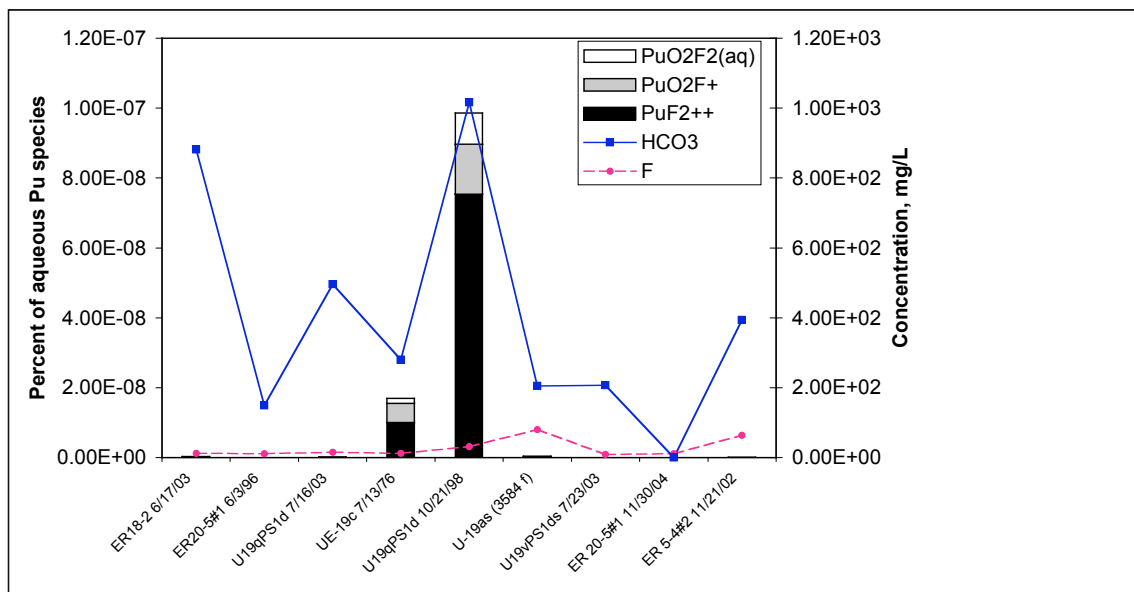


Figure 4. Aqueous plutonium-fluoride species, and HCO₃ and F concentrations in nine selected NTS wells.

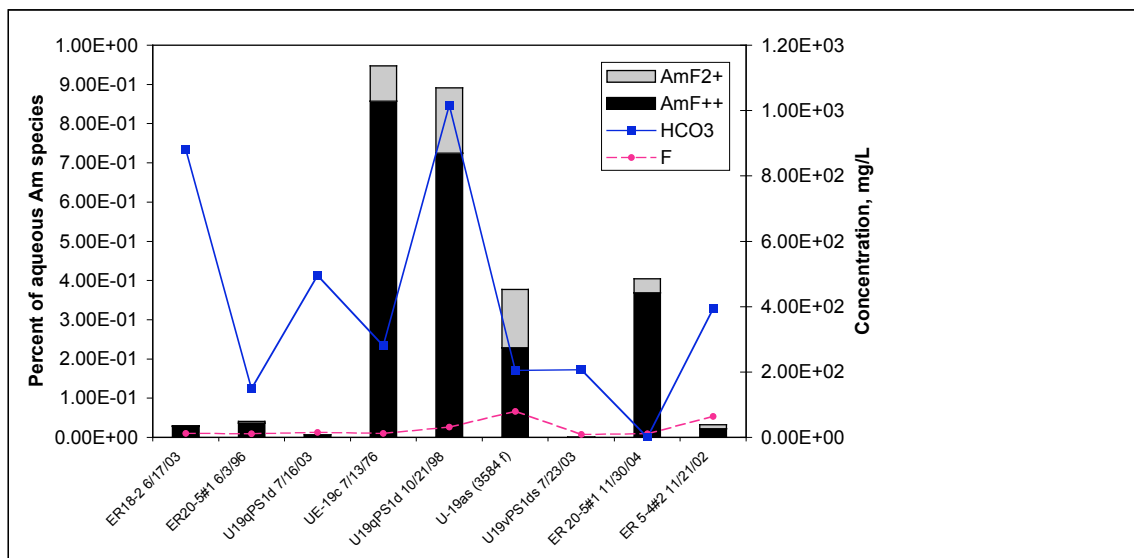


Figure 5. Aqueous americium-fluoride species, and HCO₃ and F concentrations in nine selected NTS wells.

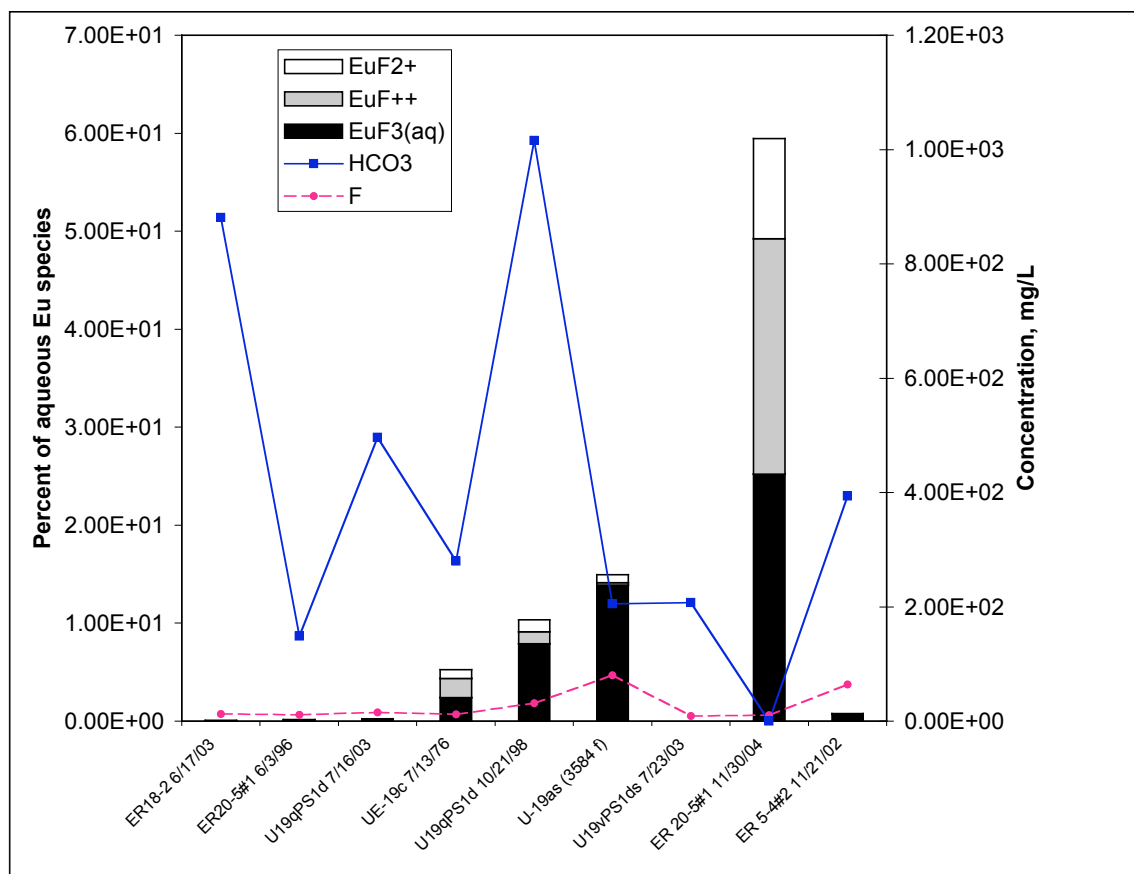


Figure 6. Aqueous europium-fluoride species, and HCO₃ and F concentrations in nine selected NTS wells.

Table 1. Compilation of chemical analyses of NTS waters with F concentrations greater than 10 mg/L. All values in mg/L except where noted.

Well name	Sample Date	Origination	pH	T°C	Cond. mS/cm	DIC	HCO ₃	CO ₃	F	Cl	Br	NO ₃	SO ₄	Na	K	Ca	Mg	SiO ₂
ER-18-2	6/17/03	T. Rose, LLNL, 1/2005	7.9	43.0	1277	881	853	3.7	12.5	12.3	<0.2	<0.2	52.9	344	2.1	5.9	0.5	---
ER-18-2	3/21/00	T. Rose, LLNL, 1/2005	7.6	55.2	1439	871	822	2.0	12.6	13.3	---	<1.0	53.0	365	1.8	6.1	0.2	---
ER-18-2	5/16/99	SNJV: ITEMS Database	8.0	---	---	---	780	12	11.0	12.0	0.23	---	58.0	266	12.6	9.2	0.0	81.9
ER-18-2	3/21/00	SNJV: ITEMS Database	7.9	---	---	---	890	60	13.0	13.0	0.12	---	55.0	350	3.7	5.4	1.0	40.6
ER-18-2	6/17/03	SNJV: DRI Electronic	7.7	---	---	---	849	0	13.1	11.4	0.12	---	56.1	366	2.4	6.1	0.2	46.8
ER-18-2	6/17/03	SNJV: SNJV Electronic	7.9	---	---	---	841	30	13.0	13.0	0.40	---	55.0	380	3.8	5.3	0.1	42.8
ER-18-2	5/16/99	UGTA SN Database	8.0	---	1340	---	390	---	11.0	12.0	0.23	---	58.0	26	12.6	9.2	1.1	---
ER-18-2	3/21/00	UGTA SN Database	---	---	---	---	---	---	13.0	13.0	0.12	---	55.0	350	3.7	5.6	---	---
ER-18-2	6/17/03	UGTA SN Database	7.9	---	1500	---	690	---	13.0	13.0	0.40	---	55.0	370	3.7	5.2	0.1	---
ER-18-2	6/17/03	UGTA SN Database	---	---	1500	---	690	---	13.0	13.0	0.40	---	55.0	380	3.8	5.3	0.1	---
ER-18-2	6/17/03	UGTA SN Database	7.7	---	1400	---	849	0	13.1	11.4	0.12	0.35	56.1	366	2.4	6.1	0.2	---
ER-20-5 #1	11/30/04	T. Rose, LLNL, 1/2005	8.3	---	546	---	---	---	10.8	24.7	0.07	1.80	43.2	118	4.6	6.2	0.1	---
ER-20-5 #1	6/3/96	SNJV: ITEMS Database	8.5	---	---	---	149	8.0	11.5	26.9	0.25	---	41.3	107	6.0	11.0	0.9	72.1
ER-20-5 #1	6/3/96	UGTA SN Database	8.5	---	516	---	---	---	11.5	26.9	0.25	---	41.3	107	6.0	11.0	0.9	---
ER-20-5 #1	4/22/97	UGTA SN Database	8.2	34.1	---	186	---	---	10.1	23.0	0.05	2.00	39.0	104	4.5	6.6	0.3	42.8
ER-20-5 #1	6/3/96	UGTA SN Database	8.4	35.0	---	187	---	---	10.3	26.4	---	---	40.6	113	4.2	6.1	0.2	61.4
ER-5-4	6/20/01	UGTA SN Database	8.7	---	2800	---	200	35	37.0	22.0	0.50	---	1600	430	41.0	6.0	0.3	---
ER-5-4 #2	11/21/02	T. Rose, LLNL, 1/2005	8.7	38.1	1249	394	381	11	63.9	51.7	<0.5	<0.1	119.4	334	4.3	0.7	0.3	---
ER-5-4 #2	10/30/02	UGTA SN Database	8.6	---	1400	---	290	22	59.0	52.0	0.40	---	120.0	310	6.8	0.6	0.2	---
ER-5-4 #2	11/21/02	UGTA SN Database	8.7	---	1400	---	300	22	61.0	52.0	0.20	---	120.0	310	7.0	0.6	0.2	---
ER-5-4 #2	11/21/02	UGTA SN Database	8.7	---	1400	---	280	31	60.0	52.0	0.20	---	110.0	320	6.8	0.6	0.1	---
NC-EWDP-1DX (deep)	5/24/99	UGTA SN Database	6.7	28.1	1857	---	---	---	11.5	50.2	0.09	---	115.0	323	6.1	41.4	10.7	---
NC-EWDP-1DX (deep)	5/24/99	UGTA SN Database	6.7	29.0	1818	---	---	---	10.6	50.6	0.09	---	111.0	331	6.5	39.0	10.7	---
NC-EWDP-1DX (deep)	5/24/99	UGTA SN Database	6.7	29.0	1818	---	---	---	10.8	48.3	0.10	---	111.0	335	6.1	39.7	10.8	---
U19ad PS1A	9/27/04	T. Rose, LLNL, 1/2005	9.4	47.0	941	---	---	---	36.0	43.5	<0.02	2.98	106.4	156	6.6	0.8	0.0	---
U-19as (3584 ft)	6/7/65	SNJV: USGS Electronic	8.4	---	---	---	205	8.0	80.0	11.0	---	---	27.0	189	5.8	1.2	---	66.0
U-19as (3584 ft)	6/7/65	SNJV: USGS Electronic	9.3	---	---	---	261	82	70.0	12.0	---	---	51.0	263	7.1	1.6	---	84.0
U-19as (3584 ft)	6/7/65	SNJV: USGS Electronic	9.3	---	---	---	174	39	80.0	19.0	---	---	26.0	200	5.5	2.0	0.1	66.0
U-19as (3584 ft)	6/7/65	UGTA SN Database	8.4	---	828	---	115	5.0	80.0	11.0	---	2.50	27.0	189	5.8	1.2	27.0	---
U-19as (3584 ft)	6/7/65	UGTA SN Database	9.3	---	1154	---	150	49	70.0	12.0	---	12.0	51.0	263	7.1	1.6	51.0	---
U-19as (3584 ft)	6/7/65	UGTA SN Database	9.3	---	889	---	122	23	80.0	19.0	---	1.00	26.0	200	5.5	2.0	26.0	---
U-19bh	3/23/04	SNJV: Bechtel Electronic	---	---	---	---	168	1.2	17.9	10.2	---	---	21.6	94	10.2	4.4	3.1	103.1
U-19bh	3/23/04	UGTA SN Database	---	---	---	8	---	---	17.9	10.2	---	---	21.6	94	8.1	4.2	3.1	---
U-19q PS#1d	7/16/03	T. Rose, LLNL, 1/2005	8.0	35.2	416	496	484	2.7	15.5	7.2	<0.03	0.50	20.8	199	9.0	5.0	0.2	---
U-19q PS#1d	10/21/98	T. Rose, LLNL, 1/2005	7.2	33.0	959	1016	898	0.8	31.5	10.4	<0.1	<0.07	29.7	342	10.3	3.2	0.0	---
U-19q PS#1d	10/20/98	SNJV: ITEMS Database	---	---	---	---	---	---	28.0	12.0	0.15	---	31.0	260	5.9	0.7	0.1	68.5
U-19q PS#1d	10/20/98	UGTA SN Database	---	---	---	---	159	---	28.0	12.0	0.15	---	31.0	260	5.9	0.7	3.1	---
U-19vPS1ds	7/23/03	T. Rose, LLNL, 1/2005	9.3	35.7	517	207	185.8	20.7	8.9	53.2	0.1	0.7	0.8	140	8.8	1.0	0.1	---
U-2bs PS #1DB (Starwort)	3/11/75	UGTA SN Database	---	---	1510	380	---	0	15.0	150	---	0.30	380.0	220	50.0	47.0	0.5	---
U-2bs PS #1DB (Starwort)	6/24/75	UGTA SN Database	---	176	1370	280	---	1.0	20.0	130	---	0.50	280.0	220	43.0	24.0	1.7	---
UE-19c Water Well	7/13/76	SNJV: DRI Electronic	7.2	---	---	---	280	---	12.0	8.0	---	---	22.0	120	0.9	3.3	0.1	42.8
UE-19c Water Well	7/13/76	UGTA SN Database	7.2	---	480	22	---	0	12.0	8.0	---	---	22.0	120	0.9	3.3	0.1	---

Table 2. Aqueous species containing F in thermodynamic database data0.ymp.R4 in chemical system represented by NTS groundwaters and radionuclides of interest.

F^-	UF_2^{++}	NpF^{+++}	PuF^{+++}	AmF^{++}	EuF^{++}
AlF^{++}	UF_3^+	NpF_2^{++}	PuF_2^{++}	AmF_2^+	EuF_2^+
AlF_2^+	$UF_4(aq)$	$NpO_2F(aq)$	PuO_2F^+		$EuF_3(aq)$
$AlF_3(aq)$	UF_5^-	NpO_2F^+	$PuO_2F_2(aq)$		
AlF_4^-	UF_6^{--}	$NpO_2F_2(aq)$			
CaF^+	UO_2F^+				
$HF(aq)$	$UO_2F_2(aq)$				
HF_2^-	$UO_2F_3^-$				
MgF^+	$UO_2F_4^{--}$				
$NaF(aq)$	UF^{+++}				
SiF_6^{--}					

Table 3. Contribution of U-F complexes to the aqueous speciation of U in selected NTS groundwaters, expressed in percent of total aqueous concentration of U.

	ER18-2 6/17/03	ER20-5#1 6/3/96	U19qPS1d 7/16/03	UE-19c 7/13/76	U19qPS1d 10/21/98	U-19as (3584') 6/7/65	U19vPS1ds 7/23/03	ER 20-5#1 11/30/04	ER 5-4#2 11/21/02
UO₂F₂(aq)	4.1E-06	2.3E-05	2.3E-05	1.0E-02	2.9E-03	5.8E-04	2.5E-08	3.9E-03	5.9E-06
UO₂F⁺	2.6E-06	1.5E-05	1.1E-05	6.1E-03	7.2E-04	5.5E-05	2.1E-08	2.6E-03	7.3E-07
UO₂F₄--	2.3E-09	1.0E-08	3.3E-06	1.1E-03	8.2E-04	4.3E-04	2.0E-09	3.9E-04	3.5E-06
UO₂F₃-	4.7E-07	2.4E-06	1.9E-08	4.7E-06	1.0E-05	1.3E-05	6.6E-12	1.5E-06	8.6E-08
UF₄(aq)	5.5E-29	2.1E-29	3.2E-28	3.4E-24	6.1E-24	3.4E-26	2.9E-34	6.9E-27	5.3E-29
UF₃⁺	1.2E-29	4.5E-30	5.3E-29	7.0E-25	5.2E-25	1.1E-27	8.2E-35	1.5E-27	2.2E-30
UF₅-	8.4E-31	3.0E-31	6.1E-30	5.1E-26	2.4E-25	3.4E-27	3.2E-36	9.2E-29	4.1E-30
UF₂⁺⁺	1.4E-31	4.8E-32	6.6E-31	4.0E-27	5.4E-26	1.8E-27	1.9E-37	6.5E-30	1.9E-30
UF₆--	7.7E-32	2.3E-32	4.5E-31	7.2E-27	2.4E-27	1.8E-30	1.2E-36	1.7E-29	5.0E-33
UF⁺⁺⁺	5.5E-35	1.6E-35	1.2E-34	2.2E-30	3.8E-31	9.1E-35	5.1E-40	5.6E-33	3.8E-37
Total concentration (mg/L)									
F	1.3E+01	1.2E+01	1.6E+01	1.2E+01	3.2E+01	8.0E+01	8.9E+00	1.1E+01	6.4E+01
HCO₃	8.8E+02	1.5E+02	5.0E+02	2.8E+02	1.0E+03	2.1E+02	2.1E+02	0.0E+00	3.9E+02

Table 4. Contribution of Np-F complexes to the aqueous speciation of Np in selected NTS groundwaters, expressed in percent of total aqueous concentration of Np.

	ER18-2 6/17/03	ER20-5#1 6/3/96	U19qPS1d 7/16/03	UE-19c 7/13/76	U19qPS1d 10/21/98	U-19as (3584') 6/7/65	U19vPS1ds 7/23/03	ER 20-5#1 11/30/04	ER 5-4#2 11/21/02
NpO₂F(aq)	1.6E-01	2.2E-01	2.7E-01	6.8E-01	1.2E+00	1.3E+00	2.6E-02	7.7E-01	3.5E-01
NpO₂F⁺	2.6E-12	9.3E-13	3.4E-12	5.3E-11	9.4E-11	6.6E-12	1.6E-14	4.7E-12	8.9E-13
NpO₂F₂(aq)	1.4E-12	5.1E-13	2.4E-12	3.0E-11	1.3E-10	2.5E-11	6.8E-15	2.5E-12	2.5E-12
NpF₂⁺⁺	8.8E-17	1.9E-18	3.4E-12	3.9E-14	2.0E-13	1.3E-16	5.6E-22	1.9E-17	3.8E-18
NpF⁺⁺⁺	4.7E-20	8.3E-22	3.1E-20	1.6E-17	4.1E-17	9.3E-21	3.3E-25	8.6E-21	3.9E-22
Total concentration (mg/L)									
F	1.3E+01	1.2E+01	1.6E+01	1.2E+01	3.2E+01	8.0E+01	8.9E+00	1.1E+01	6.4E+01
HCO₃	8.8E+02	1.5E+02	5.0E+02	2.8E+02	1.0E+03	2.1E+02	2.1E+02	0.0E+00	3.9E+02

Table 5. Contribution of Pu-F complexes to the aqueous speciation of Pu in selected NTS groundwaters, expressed in percent of total aqueous concentration of Pu.

	ER18-2 6/17/03	ER20-5#1 6/3/96	U19qPS1d 7/16/03	UE-19c 7/13/76	U19qPS1d 10/21/98	U-19as (3584') 6/7/65	U19vPS1ds 7/23/03	ER 20-5#1 11/30/04	ER 5-4#2 11/21/02
PuO₂F⁺	2.3E-10	1.5E-11	1.8E-10	5.5E-09	1.4E-08	1.5E-10	2.6E-13	3.2E-11	3.0E-11
PuO₂F₂(aq)	5.7E-11	3.9E-12	5.8E-11	1.4E-09	9.0E-09	2.5E-10	5.0E-14	7.6E-12	3.8E-11
PuF₂⁺⁺	1.9E-11	7.8E-14	1.1E-11	1.0E-08	7.5E-08	7.5E-12	2.3E-17	3.2E-13	3.2E-13
PuF⁺⁺⁺	9.2E-15	3.1E-17	3.7E-15	3.9E-12	1.4E-11	4.7E-16	1.2E-20	1.3E-16	3.0E-17
Total concentration (mg/L)									
F	1.3E+01	1.2E+01	1.6E+01	1.2E+01	3.2E+01	8.0E+01	8.9E+00	1.1E+01	6.4E+01
HCO₃	8.8E+02	1.5E+02	5.0E+02	2.8E+02	1.0E+03	2.1E+02	2.1E+02	0.0E+00	3.9E+02

Table 6. Contribution of Am-F complexes to the aqueous speciation of Am in selected NTS groundwaters, expressed in percent of total aqueous concentration of Am.

	ER18-2 6/17/03	ER20-5#1 6/3/96	U19qPS1d 7/16/03	UE-19c 7/13/76	U19qPS1d 10/21/98	U-19as (3584') 6/7/65	U19vPS1ds 7/23/03	ER 20-5#1 11/30/04	ER 5-4#2 11/21/02
AmF++	2.8E-02	3.7E-02	6.6E-03	8.6E-01	7.2E-01	2.3E-01	5.8E-04	3.7E-01	2.2E-02
AmF2+	2.5E-03	3.7E-03	4.7E-06	9.0E-02	1.7E-01	1.5E-01	4.4E-05	3.6E-02	1.0E-02
Total concentration (mg/L)									
F	1.3E+01	1.2E+01	1.6E+01	1.2E+01	3.2E+01	8.0E+01	8.9E+00	1.1E+01	6.4E+01
HCO3	8.8E+02	1.5E+02	5.0E+02	2.8E+02	1.0E+03	2.1E+02	2.1E+02	0.0E+00	3.9E+02

Table 7. Contribution of Eu-F complexes to the aqueous speciation of Eu in selected NTS groundwaters, expressed in percent of total aqueous concentration of Eu.

	ER18-2 6/17/03	ER20-5#1 6/3/96	U19qPS1d 7/16/03	UE-19c 7/13/76	U19qPS1d 10/21/98	U-19as (3584') 6/7/65	U19vPS1ds 7/23/03	ER 20-5#1 11/30/04	ER 5-4#2 11/21/02
EuF3(aq)	3.4E-02	6.6E-02	1.3E-01	2.4E+00	7.9E+00	1.4E+01	5.3E-04	2.5E+01	7.0E-01
EuF++	3.4E-02	5.9E-02	7.0E-02	2.0E+00	1.2E+00	2.8E-01	8.1E-04	2.4E+01	2.6E-02
EuF2+	1.3E-02	2.6E-02	3.8E-02	9.0E-01	1.2E+00	8.0E-01	2.7E-04	1.0E+01	5.4E-02
Total concentration (mg/L)									
F	1.3E+01	1.2E+01	1.6E+01	1.2E+01	3.2E+01	8.0E+01	8.9E+00	1.1E+01	6.4E+01
HCO3	8.8E+02	1.5E+02	5.0E+02	2.8E+02	1.0E+03	2.1E+02	2.1E+02	0.0E+00	3.9E+02

References

Grenthe, I., Fuger, J., Konings, R.J.M., Lemire, R.J., Muller, A.B., Nguyen-Trung, C. and Wanner, H. (1992) Chemical Thermodynamics of Uranium. Volume 1 of Chemical Thermodynamics. Wanner, H. and Forest, I., eds. Amsterdam, The Netherlands: North-Holland Publishing Company.

Lemire, R.J., Organisation for Economic Co-operation and Development (OECD)/ Nuclear Energy Agency (NEA) (2001) Chemical Thermodynamics of Neptunium and Plutonium. New York, New York: Elsevier.

Lemire, R.J. and Tremaine, P.R. (1980) Uranium and plutonium equilibria in aqueous solutions to 200°C. J. Chem. Eng. Data, 25, 361-370.

Rose, Timothy (2005) Personal communication of Nevada Test Site Under Ground Test Area (UGTA) water quality data for year 2004. To be included in future revisions of the Stoller-Navarro database. Lawrence Livermore National Laboratory

Silva, R.J., Bidoglio, G., Rand, M.H., Robouch, P.B., Wanner, H., and Puigdomenech, I. (1995) Chemical Thermodynamics of Americium. Volume 2 of Chemical Thermodynamics. Amsterdam, The Netherlands: Elsevier.

Spahiu, K. and Bruno, J. (1995) A Selected Thermodynamic Database for REE to be Used in HLNW Performance Assessment Exercises. SKB Technical Report 95-35. Stockholm, Sweden: Swedish Nuclear Fuel and Waste Management Company.

SNJV (2005) Excel database provided by Nicole Denovio, Stoller Navarro Joint Venture. Compiled for use in the FY05 Upscaling Radionuclide Retardation Task.

Stoller-Navarro (2004) Comprehensive Water Quality Database for Groundwater in the Vicinity of the Nevada Test Site, Revision 6. Compact Disc database with user interface. Las Vegas: Stoller-Navarro Joint Venture, SN/99205-026.

Wolery, T.J. (2002) EQ3/6 - Software for Geochemical Modeling, Version 8.0, UCRL-CODE-2003-009, Lawrence Livermore National Laboratory, Livermore, California.